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Für die Beurteilung der Patentfähigkeit in Betracht zu ziehende Druckschriften:

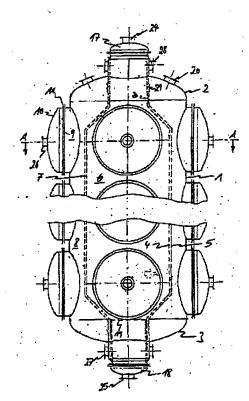
**NICHTS ERMITTELT** 

#### Die folgenden Angeben sind den vom Anmelder eingereichten Unterlagen entnommen

(A) Reaktor

Es wird ein Reaktor, der insbesondere für die Erzeugung von Synthesegas durch partielle Oxidation geeignet ist, beschrieben.

Der erfindungsgemäße Reaktor ermöglicht die Realisierung eines Reaktors mit einer nahezu beliebig großen Leistung, wobel eine ausreichende Kühlung wesentlicher und zum Teil stark druckbelasteter metallischer Bauteile realisiert werden kann. Ferner wird ein sicherer und gasdichter Übergang von dem als Membranrohr ausgebildeten Bereich eines Reaktorrohres zu den metallischen Bautellen des Reaktorrohres gewährleistet. Des Weiteren können schadhafte Reaktorrohre vergleichsweise einfach und schnell ausgewechselt werden. Darüber hinaus ist der erfindungsgemäße Reaktor so konventionell als möglich aufgebaut, so dass der Anteil der keramischen Bauteile gering gehalten werden kann. Es wird zudem eine verbesserte Reaktionsführung ermöglicht, da die Zuführung des Sauerstoffes zu dem Kohlenwasserstoff oder Kohlenwasserstoffgemisch über die Rohrlänge - d. h. entlang des Reakti nswege - erfolgen kann.



#### OffenlegungsschriftInternational Class

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Documents to be taken into consideration for evaluating the patentability: NOTHING FOUND

#### Reactor

A reactor is described which is especially suitable for the generation of synthesis gas by partial oxidation.

The reactor in accordance with the invention makes it possible to achieve a reactor with a practically arbitrarily high performance, wherein an adequate cooling of essential and in some cases heavily pressure-loaded metal components can be achieved. In addition, a safe and gas-tight transition from the area of a reactor pipe designed as a membrane pipe to the metal components of the reactor pipe is guaranteed. Furthermore, damaged reactor pipes can be replaced relatively simply and rapidly. In addition, the reactor in accordance with the invention is designed as conventionally as possible, so that the fraction of ceramic components can be kept low. In addition, an improved reaction control is made possible, since the feeding of the oxygen into the hydrocarbon or hydrocarbon mixture can take place over the pipe length, ie, along the reaction path.

#### Specification

[0001] The invention pertains to a reactor which is especially suitable for the production of synthesis gas by partial oxidation.

[0002] To produce synthesis gas, a gas-tight, oxygen ion- and electron-conducting ceramic membrane is fed on one side (retentate side) with a hot, oxygen-containing gas mixture. On the other side of the membrane (permeate side) the oxygen that emerges is reacted with a fed-in hydrocarbon or hydrocarbon mixture to form a synthesis gas.

[0003] The oxygen ion transport through such ceramic membranes, however, takes place in the desired direction only if the oxygen partial pressure on the retentate side is greater than on the permeate side.

[0004] On the permeate side of the membrane, as a result of the chemical reaction with the hydrocarbon or hydrocarbons, the partial pressure of oxygen is very low, so that the oxygen-containing gas mixture fed to the retentate side need only be compressed to a relatively low pressure. Usually the pressure of the synthesis gas formed is even greater than that of the oxygen-containing gas mixture.

[0005] The optimal working or effective range of available ceramic membranes lies at temperatures between 700°C and 1100°C.

[0006] From German patent application 100 29 883.4 a reactor is known for production of synthesis gas by partial oxidation, in which a plurality of membrane pipes suspended in a pipe base are arranged in parallel to the longitudinal axis of the reactor. By means of a suitable guidance of a cooling air stream in this reactor design it is achieved that all of the highly pressure-loaded metal components are adequately cooled.

[0007] The performance of this reactor design, however, is limited on one hand by the diameter of the container. This means that to achieve certain performances, several reactors must be connected together, which result in a high space requirement and considerable costs for the pipelines that connect the individual reactors. It is also indispensable that some fittings of this reactor must

be made from high-temperature-resistant special steel. Furthermore it is necessary as a rule that the reactor jacket, which may be exposed to a relatively high pressure load, is either provided on its inside with a thermal insulating layer to reduce the temperature level, or on the other hand likewise consists of a special steel. If the heat insulating layer is provided, a so-called outer skin temperature monitoring is also necessary.

[0008] It may also be necessary that so-called turbulence enhancers be provided in the areas of the gas space surrounding the membrane pipes.

Theoretically, in the reactor design described in German patent application 100 29 883.4 a uniform gas flow into the gas spaces can be achieved only with a relatively high technical effort because of the radial inflow and outflow of the gas streams and the relatively low ratio of height to diameter of the gas spaces.

[0009] If a granular catalyst to be packed in is used in the reactor construction described in German patent application 100 29 883.4, the filling

[0010] The goal of the present invention is to provide a reactor for the production of synthesis gas that avoids the disadvantages mentioned.

and emptying of the inner container that received the catalyst is technically

challenging and time-consuming, since the inner container must be removed for

[0011] To accomplish this task, a reactor is suggested.

#### [0012]

this purpose.

- --with a reactor jacket
- --with two caps that close off the two ends of the reactor jacket
- --with two shafts arranged one inside the other and concentric to one another in the reactor jacket, an inner shaft and an outer shaft
- --that define an inner gas space, a first annular gas space and a second annular gas space
- --wherein in each case at least two openings are assigned to the inner gas space and the first annular gas space and at least one opening is assigned to the second annular gas space

- --with at least one chamber, preferably several chambers arranged over the circumference
- --wherein each chamber has at least one opening
- --with pipe plates arranged in the chambers, with at least one removal pipe fastened in the pipe plate essentially perpendicular to the reactor jacket
  - -with at least one area of the inner shaft that is designed as a pipe plate
- --with at least one pipe fastened in the area of the inner shaft serving as the pipe plate, arranged essentially perpendicular to the reactor jacket, which is at least partially formed as a membrane pipe
- --wherein the area of the pipe or pipes formed as a membrane pipe is preferably arranged in the inner gas space
- -- and where the removal pipes are arranged coaxially to the pipes and extend into these.
- [0013] Additional advantageous embodiments of the reactor in accordance with the invention are objects of the subclaims.
- [0014] The reactor in accordance with the invention as well as additional designs thereof will be explained in greater detail on the basis of the exemplified embodiment represented in Figures 1 to 3.
- [0015] These show the following:
- [0016] A side view of a possible embodiment of the reactor in accordance with the invention.
- [0017] Figure 2. A sectional view along line A-A through the embodiment shown in Figure 1.
- [0018] Figure 3 structure and arrangement of a membrane pipe as well as a removal pipe. Such reactors—as shown in Figure 1—are generally designed in a cylindrically symmetric pattern. They can be arranged either upright or in any other alignment, for example, horizontal. In the following the upright arrangement shown in Figure 1 will be described.
- [0019] The reactor consists of a reactor jacket 1 (in the following designated only as "jacket" and two caps 2 and 3 that close off the two ends of the jacket

1. In practice the lower cap 3—unlike the representation in Figure 1—is designed merely as a plate.

[0020] Within the jacket 1, two shafts arranged one inside the other and concentric to one another—an inner shaft 4 and an outer shaft 5—are provided. These define an inner gas space 6, a first annular gas space 7 and a second annular gas space 8—also designated as hot air shafts in the following. [0021] To the inner gas space 6 and the first annular gas space 7, in the embodiment of the reactor in accordance with the invention shown in Figures 1 and 2, in each case two openings, 24, 25, 27, and 28 and to the second annular gas space 8 and opening 20 is assigned. Under the term "an opening"—as will become clear on the basis of the description that follows—an opening is defined over which a certain gas stream is conveyed to the reactor or removed from it. The fact that several openings that serve the same purpose may be present over the circumference of the pipe jacket under certain circumstances—as shown in Figures 1 and 2—is obvious.

On the circumference of the jacket 1, the chambers 9 are now [0022] arranged in several stages. These can be made in a closed design or—as shown in Figures 1 and 2-provided with removable caps 10. The chambers 9 or their caps 10, however, each have at least one opening 26. In the chambers 9, pipe plates 11, which preferably are designed to be removable, are arranged. In the pipe plates 11 in turn a plurality of removal pipes 12 are inserted or insertable. The arrangement of the removal pipes 12—as well as the pipes 14, as [0023] will be discussed in greater detail in the following-preferably takes place in layers. Both the removal pipe 12 and the pipe 14 are arranged essentially perpendicular to the jacket 1 or its longitudinal axis. Through this arrangement of the aforementioned pipes, their length no longer has any limiting effects on the performance of the reactor. Because of the possibility of arranging a plurality of layers of pipes one atop the other or in the direction of the longitudinal axis, a high capacity can also be achieved with a single tower-type reactor.

[0024] The pipe plates 11 preferably have pipe pieces 13 which are welded into the pipe plates 11 and into which the removal pipes can be inserted. The removal pipes 12 inserted in this way are welded tightly together with the pipe pieces 13. If it is necessary to replace defective removal pipes 12, these can be removed from the pipe piece 13 after removal of the weld seam. However, welding is not absolutely necessary, since under certain circumstances it is possible to entirely avoid a fixed connection, or connecting methods that are alternative to welding may be employed.

[0025] The previously described inner shaft 4 is partially designed as a pipe plate for the reactor pipes 14. The number of these [pipes] corresponds to the number of removal pipes 12. The removal pipes 12 are arranged coaxially to the pipes 14 and extend into these. The pipes 14 are in turn preferably welded into the inner shaft 4 and are inserted or insertable into pipe pieces 15 welded to the outer shaft 5. In terms of the connecting methods between the pipe pieces [sic] and the pipes 14, reference may be made to the abovementioned statements.

[0026] The reactor pipes 14 are at least partially designed as membrane pipes 16. The area of the pipes 14 designed as the membrane pipes 16 is preferably arranged exclusively in the inner gas space 4.

[0027] The removal pipes 12 extend into the interior of the reactor pipe 14 and preferably over the total area of the pipe 14 designed as a membrane pipe 16.

[0028] The area of the reactor pipe 14 designed as a membrane pipe 16 can be designed either in the form of a gas-tight, oxygen ion- and electron-conducting ceramic membrane applied on a gas-permeable support pipe or in the form of a pipe consisting of a monolithic, gas-tight ceramic that is conductive for oxygen ions and electrons.

[0029] As shown in Figure 3, the area of the pipe 14 designed as a membrane pipe 16 is preferably connected in a material-locking and coaxial manner at both of its ends with one metal pipe each of approximately the same diameter.

The pipes 14 are fixed in the pipe pieces 15 at only one of their ends, while the opposite end to be sure is closed gas tight, but is freely expandable in the axial direction to avoid stresses due to differences in thermal expansion. The same is true for the removal pipes 12, which likewise are fastened only at one of their ends in the pipe plate 11 or the pipe pieces 13 arranged on it. Not shown in Figure 3 is a suitable support of the freely expandable end of the pipes 14. [0030] The inner shaft 4 and thus the inner gas chamber 6, according to an advantageous embodiment of the reactor in accordance with the invention, are filled at least in the area of the pipes 14 with a preferably granular catalyst material. For filling or emptying, the shaft 4 is extended in design at both ends of the reactor and provided with an upper cap 17 and a lower cap 18. At the lower end of the shaft 4 in addition a supporting grid 19 for the catalyst is arranged.

[0031] As an alternate or supplement to this, the reactor pipes 14, at least the areas of the pipes 14 designed as a membrane pipe 16, can be filled with the catalyst.

[0032] On the basis of Figure 1, the area of the reactor in accordance with the invention or the exemplified embodiment shown in Figures 1 and 2 will be explained in further detail.

[0033] The hot, oxygen-containing gas mixture is conveyed to the reactor in accordance with the invention over the opening(s) 20 in the gas chamber 8. This gas mixture, at a pressure of for example 1.5 bar, has a temperature of 900°C. For example, the production of such a gas mixture can take place in a combustion chamber under an excess of fresh air. From the gas chamber 8 this gas mixture enters the annular space 23 formed by the removal pipes 12 and the reactor pipes 14 (see Figure 3).

[0034] The gas mixture flows along the area of the reactor pipes 14 formed as a membrane pipe 16. In this process, through the membrane pipe 16, pure oxygen enters the inner gas space 6 in which the catalyst is located. To the inner gas space 6 and shaft 4, over the opening 24 a hydrocarbon mixture—

possibly with addition of steam—is conveyed with a temperature of 500°C and a pressure of about 30 bar. The hydrocarbon mixture reacts as a result of the catalyst with the oxygen to form a synthesis gas, which leaves the reaction shaft 4 over the opening 25. The synthesis gas produced at a pressure of about 30 bar has a temperature of about 950°C.

[0035] Over the removal pipes 12, the chambers 9, and the openings 26 provided in their caps 10, a gas mixture of reduced oxygen content is withdrawn from the reactor in accordance with the invention and possibly conveyed for further energy utilization.

[0036] To cool the inner shaft 4, over the opening(s) 27, fresh air is conveyed to the annular gas space 7; at a pressure of 1.7 bar this has a temperature of 90°C. The forcibly heated fresh air is withdrawn over the opening(s) 28 back from the gas space 7 and possibly the previously mentioned combustion chamber for the purpose of heating the oxygen-containing gas mixture.

[0037] Instead of fresh air, for cooling if desired steam can be introduced over the opening(s) 27 and removed over the opening(s) 28.

[0038] To compensate for the different axial thermal expansions of the inner shaft 4, the outer shaft 5, and the jacket 1, compensation means are provided in the form of compensators 29 and 30. However, the axial thermal expansions are low if—corresponding to an advantageous design of the reactor in accordance with the invention—the shaft 4 and the jacket 1 on the inside and the shaft 5 on the outside are provided with a thermal insulating layer.

[0039] Such a thermal protective insulation—including that of the caps 2 and 3—also serves to keep the temperature of the jacket 1 and the caps 2 and 3 at a relatively low level.

[0040] In the reactor design in accordance with the invention furthermore the higher pressure preferably prevails on the outside of the pipes 14. This is advantageous, since generally the compressive strength of ceramic is higher than its tensile strength.

[0041] The reactor in accordance with the invention creates a design in which the length of the membrane pipe no longer has any limiting influence on the performance of the reactor. Because of the high gas velocities achieved and the pipes with their transverse flow, high heat and materials transfer rates are guaranteed, so that the use of turbulence enchancers is not required. A uniform gas flow over the total flow cross-section can be guaranteed in the aforementioned gas spaces without increased technical investments. The filling and emptying of the inner shaft 4, which accommodates the catalyst, no longer requires any great technical or time investment, since this assembly of the reactor can be avoided.

[0042] In addition, the reactor in accordance with the invention makes it possible to achieve adequate cooling of the essentially and partially highly pressure loaded metal constituents. In addition, a reliable and gas-tight transition from the area of a pipe 14 designed as a membrane pipe to the metal components of the pipes 14 is guaranteed. Furthermore, damaged pipes can be replaced relatively simply and rapidly. In addition, the reactor in accordance with the invention is designed as conventionally as possible, so that the fraction of ceramic components can be minimized. Furthermore, an improved reaction control is made possible, since the feeding of the oxygen into the hydrocarbon or hydrocarbon mixture can take place over the length of the pipe, ie, along the reaction pathway.

[0043] In addition to the membrane pipes mentioned, the reactor in accordance with the invention is also suitable for the use of other membranes that can be integrated into the reactor design in the method and manner described.

#### Claims

#### 1. Reactor

With a reactor jacket (1), with two caps (2, 3) closing the two ends of the reactor jacket (1),

With two shafts arranged one inside the other and concentrically to one another in the reactor jacket (1)—an inner shaft (4) and an outer shaft (5) which define an inner gas space (6), a first annular gas space (7), and a second annular gas space (8),

Wherein each of the inner gas space (6) and the first annular gas space (7) has at least two openings (24, 25, 27, 28) assigned to it and a second annular gas space (8) has at least one opening (20) assigned to it,

With at least one chamber (9) preferably several chambers arranged over the circumference of the reactor jacket (1),

Wherein each chamber (9) has at least one opening (26),

With pipe plates (11) arranged in the chambers (9),

With at least one removal pipe (12) fastened in the pipe base (11) and arranged essentially perpendicular to the reactor jacket (1),

With at least one area of the inner shaft (4) which is designed as a pipe plate With at least one pipe (14) fastened in the inner area of the inner shaft (4) designed as a pipe plate, arranged essentially to the reactor jacket (1), which is designed at least partially as a membrane pipe (16),

Wherein the area designed as a membrane pipe (16) of the pipe or pipes (14) is preferably arranged in the inner gas space (6)

And wherein the removal pipes (12) are arranged coaxially to the pipes (14) and extend into these.

- 2. Reactor in accordance with claim 1, characterized in that the area of the pipe (14) designed as a membrane pipe (16) is designed in the form of a gas-tight, oxygen ion- and electron-ion-conducting ceramic membrane applied to a gas-permeable support pipe.
- 3. Reactor in accordance with claim 1, characterized in that the area of the pipe (14) designed as a membrane pipe (16) is designed in the form of a

- pipe consisting of a monolithic, gas-tight, oxygen ion- and electronconducting ceramic.
- 4. Reactor in accordance with one of the preceding claims, characterized in that the inner gas space (6) is at least partially filled with a preferably granular catalyst.
- 5. Reactor in accordance with one of the preceding claims, characterized in that the pipe (14), at least the area of the pipe (14) designed as a membrane pipe (16), is filled with a catalyst.
- 6. Reactor in accordance with one of the preceding claims, characterized in that on the inside of the inner shaft (4), the outside of the outer shaft (5), and/or on the inside of the separator jacket (1), a thermally insulating layer is arranged.
- 7. Reactor in accordance with one of the preceding claims, characterized in that on the pipe plate or plates (11) and/or the area of the inner shaft (4) formed as a pipe plate, pipe pieces (13, 15) are arranged, to which the pipe (14) and/or the removal pipe (17) can be fastened.
- 8. Separator in accordance with one of the preceding claims, characterized in that means for compensating for the axial thermal expansion of the inner shaft (4) and/or the outer shaft (5) are provided.
- 9. Reactor in accordance with one of the preceding claims, characterized in that the reactor is designed in a cylindrically symmetrical fashion.

Two (2) pages of drawings attached.

